Theoretical Studies of Time Dependent/Independent Radiative Transfer Including Inelastic Scattering for Both Active and Passive Sources

George W. Kattawar
Dept. of Physics
Texas A&M University
College Station, TX   77843-4242
phone: (409) 845-1180   fax: (409) 845-2590   email: kattawar@tamu.edu

LONG-TERM GOALS

We wish to develop the theoretical and computational groundwork for a new and innovative program for the remote detection and characterization of both organic and inorganic aerosols using both active (lidar) and passive techniques. This aerosol study should be very beneficial to the Navy in the areas of communications, high power laser transmission, air-sea interactions, and standoff biological detection methods. We also want to continue our collaborative program in polarimetry with the group in Minsk headed by Dr. Eleonora Zege.

OBJECTIVES

We will incorporate the full Mueller matrix formulation in this study to extract *everything* optically that can be extracted from the scattering and fluorescing aerosols. This study will also include the use of state-of-the-art techniques to calculate the single scattered Mueller matrix (SSMM) for both single particles as well as ensembles of particles that have different morphology and optical properties. We will show how to extend the SSMM into an "effective" multiple scattering Mueller matrix (MSMM) when multiple scattering has to be taken into account. We will also show that by using the MSMM it may be possible to not only determine particle optical properties and number densities but particle morphology as well. We will then determine which Mueller matrix elements or combination of them are most effective for unique aerosol signatures.

APPROACH

The success of this study for an active system depends on having programs to accurately calculate the complete time-resolved Mueller matrix for a realistic source-receiver geometry for a medium consisting of atmospheric gasses, clouds, and aerosols. We have already developed very powerful Monte Carlo programs that will calculate the multiple scattering Mueller matrix for any inhomogeneous atmosphere-ocean system which may contain a stochastic surface such as a dielectric interface separating the atmosphere from either a body of water or solid surface. These programs must be capable of handling not only backscattering (monostatic configuration), which is the most common mode of operation of lidar systems, but must also be able to handle any bistatic situation where source and receiver are at positions which will optimize the distinguishing feature of the aerosol under study.

We have modified our existing state-of-the-art Monte Carlo codes to be able to handle both time dependent and time independent active sensing. For the lidar codes we now have the flexibility to have any source-receiver geometry with arbitrary field of view (FOV) for the detector. This project

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1. REPORT DATE **30 SEP 2003**	2. REPORT TYPE	3. DATES COVERED **00-00-2003 to 00-00-2003**
4. TITLE AND SUBTITLE **Theoretical Studies of Time Dependent/Independent Radiative Transfer Including Inelastic Scattering for Both Active and Passive Sources**		5a. CONTRACT NUMBER
		5b. GRANT NUMBER
		5c. PROGRAM ELEMENT NUMBER
6. AUTHOR(S)		5d. PROJECT NUMBER
		5e. TASK NUMBER
		5f. WORK UNIT NUMBER
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) **Dept. of Physics,,Texas A&M University,,College Station,,TX, 77843**		8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)		10. SPONSOR/MONITOR'S ACRONYM(S)
		11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited

13. SUPPLEMENTARY NOTES

14. ABSTRACT
We wish to develop the theoretical and computational groundwork for a new and innovative program for the remote detection and characterization of both organic and inorganic aerosols using both active (lidar) and passive techniques. This aerosol study should be very beneficial to the Navy in the areas of communications, high power laser transmission, air-sea interactions, and standoff biological detection methods. We also want to continue our collaborative program in polarimetry with the group in Minsk headed by Dr. Eleonora Zege.

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a REPORT **unclassified**	b ABSTRACT **unclassified**	c THIS PAGE **unclassified**	**Same as Report (SAR)**	**8**	

has been carried out by Mr. Deric Gray who will be receiving his Ph.D. on this research in December of 2003.

In order to run our Monte Carlo program, we still need the SSMM for the aerosols and other constituents comprising the atmospheric system. The actual experimental Mueller matrix data for aerosols of different shapes and optical properties are quite sparse at best, therefore we will have to develop our own computer codes to give us this data. Many aerosols whether from soils, minerals or organics can be highly nonspherical with large aspect ratios. Trying to model them with any type of equivalent sphere approximation can lead to substantial errors as has been pointed out by Mischenko et al.[1,2] There have been many techniques introduced to solve the problem of electromagnetic scattering from irregular particles (for an excellent summary of the methods the reader should see Mischenko, et al.[3]) the most notable of which are the discrete dipole approximation (DDA), the T-matrix approach, and the finite difference time domain method (FDTD). We have a good deal of experience with the DDA; however, even though it is capable of modeling any shape, it is quite limited to size parameters usually less than ten and also suffers from lack of accuracy in the Mueller matrix elements involving phase information. The T-matrix approach is very powerful and is able to achieve size parameters in excess of one hundred; however, it is not well suited for particles with sharp corners and edges where an inordinately large number of vector spherical wave functions may be needed. The FDTD method has a long and rich history beginning with the work of Yee[4]. This method is applicable to any particle shape and inhomogeneity as well as handling any type of incident beam The application of this method to effectively calculate light scattering properties from dielectric aerosols at a single frequency with a variety of shapes has been carried out by Yang and Liou[5]; however, virtually all comparisons were done on phase functions, the (1,1) element of the Mueller matrix, and is the least sensitive to numerical errors compared to all the other Mueller matrix elements. Since we will be interested in all the elements of the Mueller matrix, this may require modifying the type of boundary condition imposed (a crucial element in the accuracy of the FDTD) as well as a refinement of the discretization methods to yield the type of accuracy we need to produce the complete Mueller matrix. We want to extend the FDTD method to handle both dispersive and tensor permittivities as well. The reason being is that if one is using short pulsed sources the frequency spectrum can be quite wide and in most instances the permittivity will be a function of frequency and this behavior must be accounted for. Also most organic molecules and substances are optically active and therefore the permittivity will no longer be a scalar but will become a tensor. Both of these additions will involve extensive modification of the FDTD method as it presently exists.

These projects are being worked on by Pengwang Zhai and Chanhui Li both of whom are Ph.D. students in the physics department of TAMU

WORK COMPLETED

a) We have completed a study of the effect of the shape of the volume scattering function on the detection of targets in turbid media. A talk on this research was presented at the Ocean Optics XVI conference in November, 2002 and a publication on it has been accepted and will soon appear in Applied Optics (2003)

b) I have assisted the Quantum Optics Group in completing a study of a new way to detect bacterial spores by a technique which we have labeled FAST CARS (Femtosecond Adaptive Spectroscopic Techniques for Coherent Anti-Stokes Raman Spectroscopy). This article appeared in the Proceedings of the National Academy of Sciences (2003)

c) We have a completed a study of the sensitivity of the backscattering Mueller matrix to particle shape and thermodynamic phase and a paper on this has recently appeared in Applied Optics (2003)

d) We have a completed a study of the Geometric optics solution to light scattering by droxtal ice crystals and a paper on this has just been accepted for publication in Applied Optics (2003)

e) We have completed a study on the proper choice of Cartesian grid configurations for applying the FDTD method to electromagnetic scattering by dielectric particles and a paper on this has just been submitted for publication to Applied Optics (2003)

RESULTS

a) We have shown that FAST CARS can be used to generate maximal quantum coherence in large molecules and biomolecules. We have proposed that this may be a very rapid method to detect DPA (dipicolinic acid) which is common to most bacterial spores including anthrax Since each different type of spore would have its own unique mixture of metals and amino acids, it may be the case that the finer details of the Raman spectra would contain spore specific fingerprints.

b) We have shown that hexagonal ice crystals with three aspect ratios (plates, compact columns, and columns) have Mueller matrices which differ significantly from water spheres. In particular, we have shown that the (2,2) and (3,3) elements are quite sensitive to particle aspect ratio. From these results we see how the backscattering Mueller matrix can b used to discriminate between ice and water clouds.

c) A multi-faced crystal habit called a droxtal is a good representation of the small ice crystals that occur in cirrus clouds. We have used an improved geometric optics (IGO) model to calculate the Mueller matrix for both 18 and 20 faced droxtals. We have shown that there is a notable difference in the linear depolarization between hexagonal ice crystals and droxtal ice crystals at backscattering angles. This difference is also observed between the 18-faced and 20-faced crystals

d) To show the versatility of our FDTD code, we have calculated the complete optical properties for some complex particle shapes that are shown in Fig. 1. These are actual shapes of ice crystals found in cirrus clouds.

e) We have also calculated the complete Mueller matrix for bullet-rosette ice crystals which also occur in cirrus clouds. In Fig. 2 we show the geometry of these crystals with 1, 3, 4, 5, 6, 8, 9, 10, and 12 branches. In Fig. 3 we show the non-zero elements of the scattering phase matrix for bullet-rosettes with 1, 6, and 12 branches for size parameter X= 10,where $X=2\pi D/\lambda$ and λ is the wavelength. The non-zero phase matrix elements of the spheres with the same volume as the 12-branched bullet rosettes are also shown. The nonsphericity effect on the phase-matrix elements is evident from these two diagrams. It has been suggested that P_{22}/P_{11} (where P_{ij} is the ij-th element of the Mueller matrix) is indicative of nonsphericity.[6,7] For a sphere, this ratio is unity. From Fig. 3, we see the nonsphericity in terms of the deviation of P_{22}/P_{11} from unity and this effect becomes more significant with increasing branch number. The other phase matrix elements for bullet rosettes are also quite different from those for spheres. This is an extremely important result since it clearly shows that using spheres to emulate real particles can lead to quite erroneous results. We note that the elements P_{12}/P_{11}, P_{33}/P_{11}, P_{43}/P_{11}, and P_{44}/P_{11} are not particularly sensitive to the number of branches in bullet-rosettes.

3
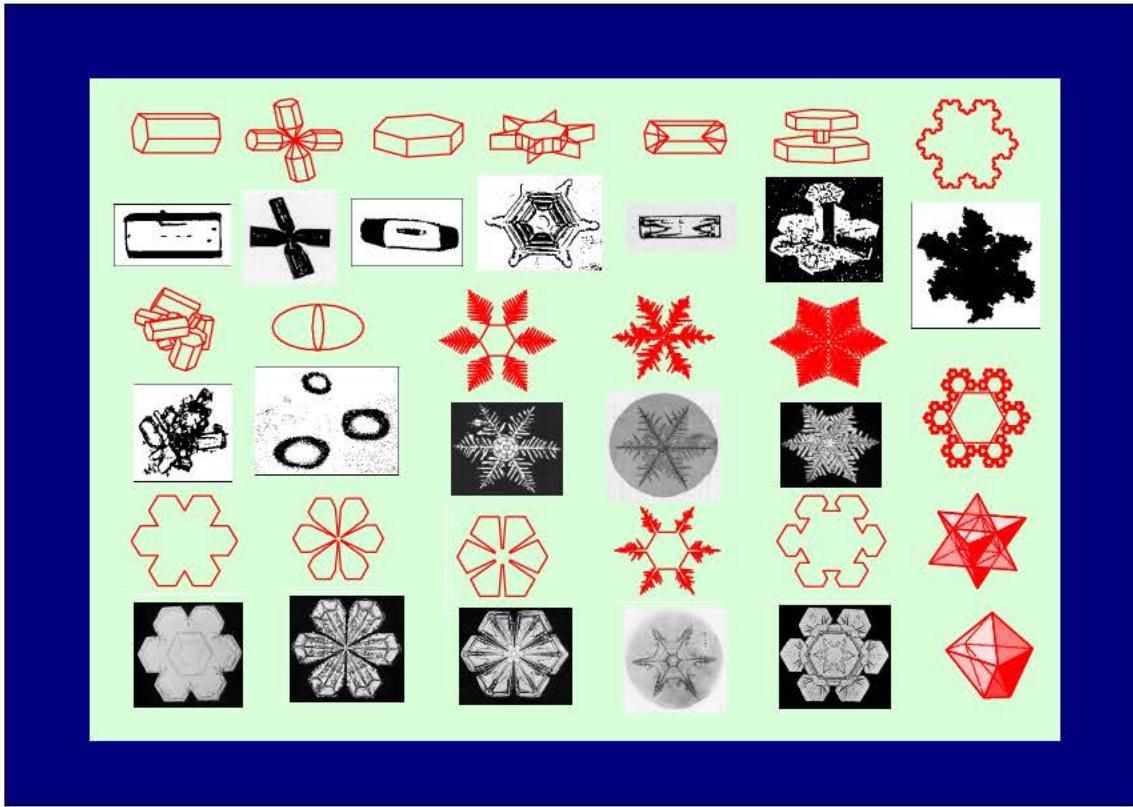
Fig. 1 A sample of the complex geometries of real particles that we can simulate in light scattering calculations with the FDTD and the geometric optics scattering-computational codes. The shapes in red are numerically defined whereas the images in black and white are from observations.
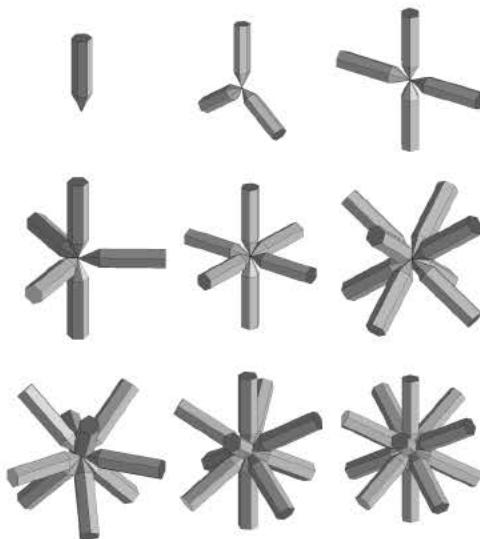
Figure 2. Morphological geometry of bullet-rosette ice crystals defined for scattering calculations.

$\lambda=0.66\ \mu m,\ x=10$
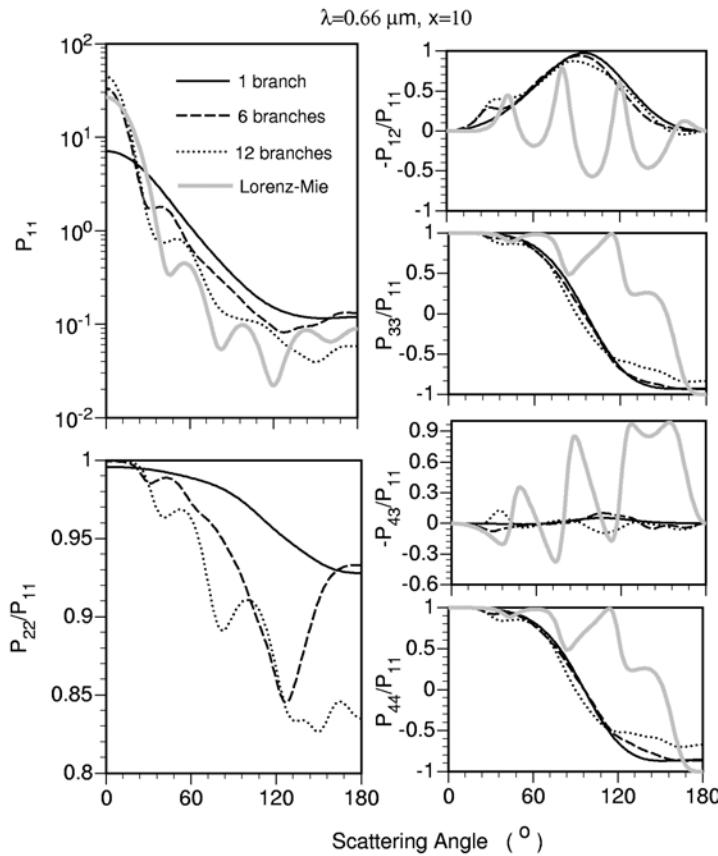
Fig. 3 Nonzero phase matrix elements for the bullet-rosette ice crystals with 1, 6 and 12 elements at 0.66 μm wavelength. The size parameter is X=5. The phase matrix elements of a sphere that has the same volume as the 12-branched bullet-rosettes are also shown.

IMPACT/APPLICATION

We now feel that from some of our preliminary work on Mueller matrix imaging, we will have new and more powerful tools for remotely sensing the atmosphere-ocean system. We also believe that it will lead to better and more comprehensive target detection in turbid media.

Our Mueller matrix work using both the FDTD and IGO (improved geometric optics method) on ice crystals has opened up a new and exciting way to remotely sense cirrus clouds and determine the nature of the ice crystals.

Another very significant application of the FDTD method is that we are now able to emulate the scattering patterns from any shape of bioaerosol. By looking at the backscattering Mueller matrix patterns from individual bioaerosol particles,we feel we may have a very rapid way to uniquely detect and identify them.

TRANSITIONS

Our Monte Carlo Mueller matrix program is presently being used by Dr. Walt McBride of PSI (Planning Systems Inc.) to calculate on a pixel-by-pixel basis the complete Mueller matrix signal received from an arbitrary object including thermal emission. This project was recently funded by the

Air Force and supports Dr. Hatcher Tynes who is now a Postdoc working with both Walt McBride and me.

RELATED PROJECTS

Our Monte Carlo code has been modified and used in a joint project with Dr. E. S. Fry to emulate a new scattering and absorption meter for the ocean. The initial results look very promising and a prototype instrument is now being built in Dr. Fry's laboratory.

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